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MEMORANDUM REPORT ARBRL-MR-02920

ASSESSMENT OF THE JAPANESE
WEAR-REDUCING ADDITIVES

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J. Richard Ward
Eli Freedman

May 1979

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US ARMY ARMAMENT RESEARCH AND DEVELOPMENT COMMAND
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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER MEMORANDUM REPORT ARBRL-MR-02920	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) "Assessment of the Japanese Wear-Reducing Additives"		5. TYPE OF REPORT & PERIOD COVERED Memorandum Report
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) J. Richard Ward Eli Freedman		8. CONTRACT OR GRANT NUMBER(s)
9. PERFORMING ORGANIZATION NAME AND ADDRESS USA Ballistic Research Laboratory ATTN: DRDAR-BLP Aberdeen Proving Ground, MD 21005		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS RDT&E 1L162618AH80
11. CONTROLLING OFFICE NAME AND ADDRESS USA Armament Research & Development Command USA Ballistic Research Laboratory ATTN: DRDAR-BL Aberdeen Proving Ground, MD 21005		12. REPORT DATE MAY 1979
		13. NUMBER OF PAGES 26
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		15. SECURITY CLASS. (of this report) Unclassified
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; distribution unlimited.		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Japanese Additives 90mm Tank Gun Alanine Wear-Reducing Additives Calcium Pyroglutamate Gun Barrel Erosion		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) meg/srf Yamazaki and co-workers at the Technical Research and Development Institute of the Japan Defense Agency reported that the addition of five percent alanine or calcium pyroglutamate to gun propellant markedly reduced the erosion rate of the gun propellant. Alanine and calcium pyroglutamate are now denoted the "Japanese additives". (continued on reverse side)		

20. ABSTRACT: (Cont'd)

Thermochemical calculations were made at BRL on the propellant the Japanese investigators tested in the 90mm tank cannon. The calculations revealed that the addition of alanine or calcium pyroglutamate reduced the flame temperature by 300 to 400K from the initial 3000K; in addition, the propellant's impetus was reduced by nearly five percent. Hence, the alanine and calcium pyroglutamate reduce erosion simply because the propellant has been diluted with a cool-burning material. Similar results could be achieved more simply by reducing the nitroglycerin or nitroguanidine content. An identical test was performed earlier in the US when five percent by weight titanium dioxide was added to M30 propellant. The TiO₂-modified propellant reduced the wear rate of the 105mm M68 tank cannon by a factor of fourteen, but sufficient volume was unavailable in the cartridge case for the modified M30 propellant.

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1. INTRODUCTION

An English translation recently appeared of a Japanese report¹ summarizing efforts at the Technical Research and Development Institute of the Japanese Defense Agency to reduce gun barrel erosion. Yamazaki and co-workers reported that addition of five percent alanine or calcium pyroglutamate to M30 propellant significantly lowered the erosivity of M30 propellant based on results in a 90mm M41 tank cannon. Since the M30 series of propellants is in the propelling charges for the four wear-limited fielded Army cannons (105mm M68, 155mm M185 and M199, and 8-inch M201), great interest has been stirred as to the utility of the Japanese additives to increase the wear life of such cannons.

In order to assess the potential of the Japanese additives, the Japanese wear data were analyzed and a series of calculations was made with the BLAKE code² to estimate the adiabatic flame temperatures and the impetuses of the propellants with alanine or calcium pyroglutamate. One explanation for the reduced erosivity of the modified M30 propellant is that the alanine or calcium pyroglutamate simply lowers the flame temperature. The effect of propellant flame temperature on barrel erosion is illustrated in Figure 1 with results from a Naval 5"/54 gun.³ In addition to reducing flame temperature, the alanine or calcium pyroglutamate will reduce propellant impetus which means more propellant will be needed to achieve the same ballistics. If the impetus is appreciably smaller than the impetus for M30, then the modified M30 propellant is no more novel an approach to reducing erosion than reducing the nitroglycerin or nitroguanidine content of M30.

II. 90MM TANK CANNON EROSION RESULTS

The modified M30 propellants were evaluated in Japan in a wear test with three 90mm tank cannons. The cannons were fabricated specifically for the wear test and had a 0.03-0.05mm chromium plate on the barrel surface. The wear test consisted of daily fifty-round bursts of M431 HEAT-T cartridges fired at intervals of thirty seconds. Muzzle velocity, chamber pressure, and accuracy were taken from the first ten to twelve rounds fired each day. Pullover gage measurements were made at the start and at the end of each day's firing.

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1. H. Yamazaki, "Study on Reducing Gun Tube Erosion", JDA Technical Research and Development Institute Report, undated.
 2. E. Freedman, "BLAKE - A Ballistic Thermodynamic Code Based on TIGER", *Proceedings of the International Symposium on Gun Propellants*, October 1973.
 3. M.C. Shamblen, "Overview of Erosion in US Naval Guns", *Proceedings of the Tri-Service Gun Tube Wear and Erosion Symposium*, March 1977.

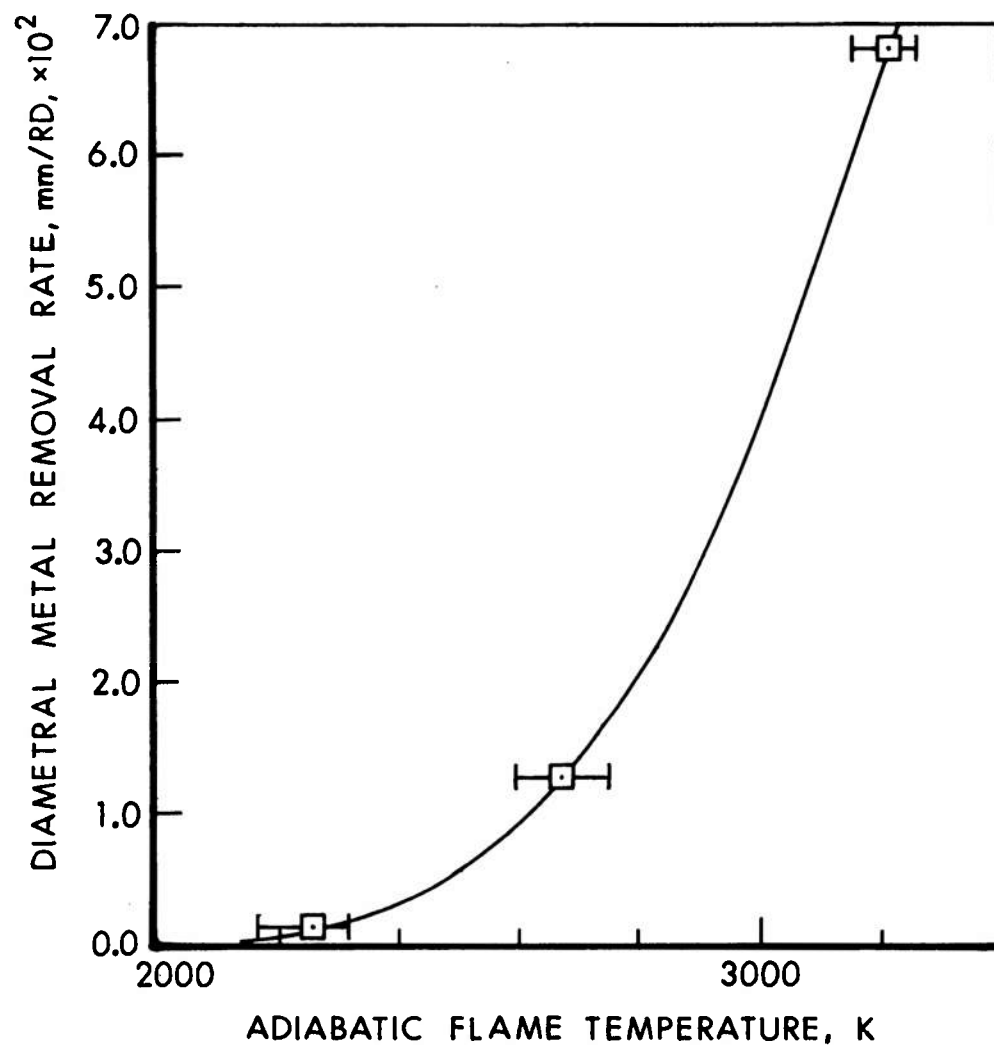


Figure 1. Results From 5"/54 High Pressure Erosion Test

The muzzle velocity drop after three 50-round bursts was 51 m/s for the standard rounds, 42 m/s for the alanine propellant, and 27 m/s for the propellant with calcium pyroglutamate. No mention is made of the initial velocity of the rounds with the Japanese additives, or whether charge weights were increased with the modified propellants.

The wear data available from the 90mm tank cannons are summarized in Table I. It is clear that the modified propellants reduce the

TABLE I. Summary of Wear Data for the 90mm Tank Cannon

<u>Rounds Fired</u>	<u>Bore Diameter Increase, mm</u>		
	<u>M30</u>	<u>Alanine</u>	<u>Ca Pyroglutamate</u>
150	2.51	0.79	0.74
200		1.62	1.62
325		3.20	

erosion rate of the 90mm tank cannon. To compare the relative erosivity of the standard M30 propellant and M30 with Japanese additive, one must separate the relative number of rounds to remove the chromium plate from the erosion of the underlying steel. Cooler propellants extend the wear life of a chromium-plated barrel in two ways. First, the number of rounds that must be fired before the chromium plate flakes or spalls from the steel substrate is greater the lower the flame temperature of the propellant. This is illustrated in Figure 2 with the number of rounds fired to remove 0.13mm chromium plate. As long as the chromium plate is intact on the bore, no wear takes place. The second way barrel life is extended is that the underlying steel erodes slower with the cooler propellant. The comparison of bore diameter increase after a certain number of rounds fired in a chromium-plated tube gives an erroneously low value compared to the bore diameter increase after firing the same number of rounds in an unplated cannon. Fortunately, the Japanese investigators provided a plot of bore erosion vs rounds fired which is reproduced as Figure 3. One sees the modified propellants increase the number of rounds that can be fired before the chromium plate is removed; the modified propellants also reduce the erosion rate of the underlying steel. To compare the relative erosivity of the modified propellants and M30 propellant on steel properly, Figure 3 shows one should compare the wear rate after fifty rounds are fired with the M30 propellant and after one hundred rounds are fired with the Japanese additives. The erosion rates are then as follows:

<u>Propellant</u>	<u>Wear Rate, mm/rd</u>
M30	0.025
Alanine Addition	.017
Calcium Pyroglutamate Addition	.018

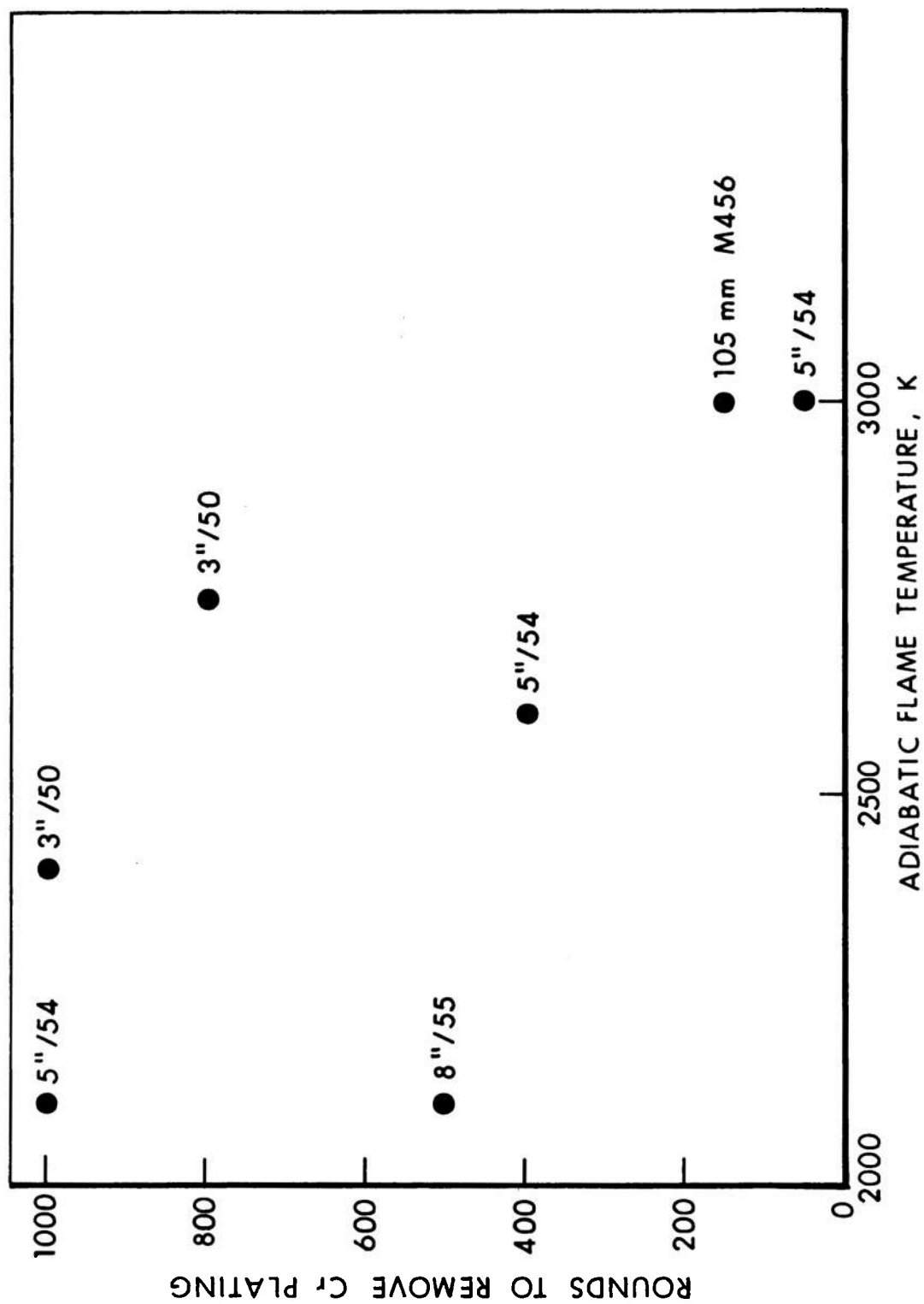


Figure 2. Number of Rounds to Remove 0.13mm Chromium Plate from Gun Barrels

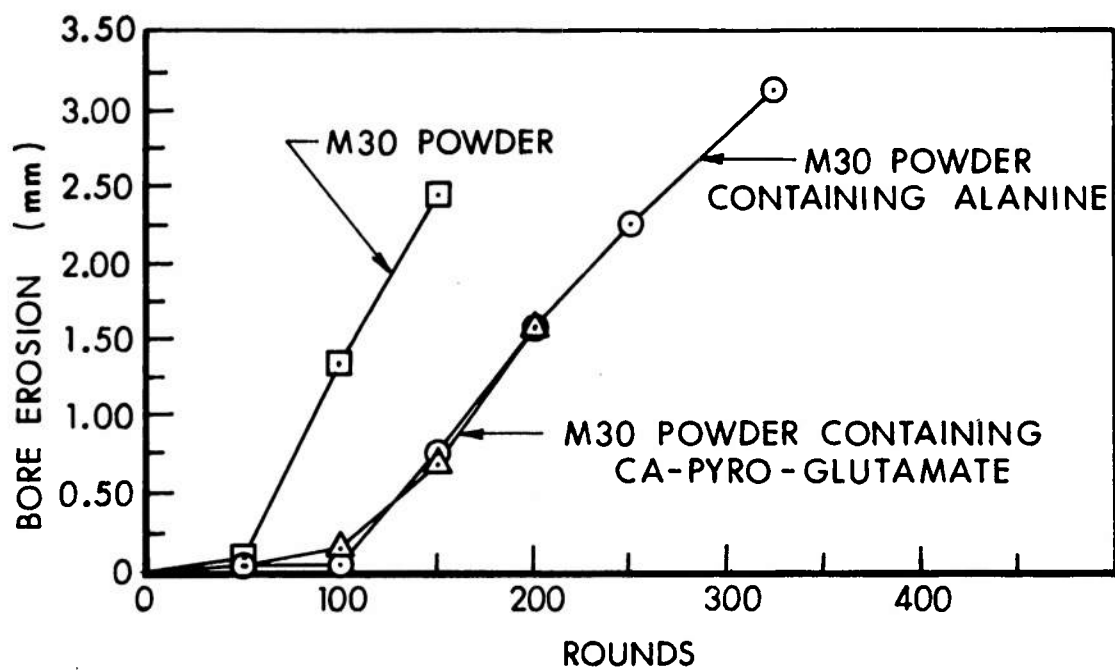


Figure 3. Bore Erosion of 90mm Gun Tubes

The modified M30 propellants reduce the wear rate of steel appreciably, but the rate of wear is twice as high as that predicted when the removal of chromium plate is not considered properly. The Japanese workers contended the modified propellant reduces the wear rate by a factor of three. In fact, the wear rate was decreased by a factor of 1.5 and the chromium plating remained intact 50 rounds longer. Using the condemnation limit of 5.00mm⁴, one sees that the wear life of the 90mm tank gun was increased from 250 rounds to 400 rounds.

The data from Table I also show that the tube wear for the propellant with calcium pyroglutamate is the same as for the propellant with alanine. This suggests that no insulating layer is formed on the bore surface with the calcium pyroglutamate propellant that might enhance the erosion reduction. The superiority of the TiO₂/wax liner has been shown to be due in large part to an insulating layer on the bore surface⁵⁻⁷, which is largely composed of TiO₂.⁸ A similar insulating layer of calcium oxide might have formed with the calcium pyroglutamate-seeded propellant; the data suggest it did not.

III. THERMOCHEMICAL CALCULATIONS

The propellant description sheet for the propellants with the Japanese additives is listed in Table II. The description sheet was taken from a subsequent report⁹ on the long-range stability of the Japanese propellants.

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4. *Evaluation of Cannon Tubes*, TM-9-1000-202-25, November 1969.
 5. F.A. Vassallo, "Heating and Erosion Techniques Applied to the Eight-Inch Howitzer", Volume I, 12th JANNAF Combustion Meeting, CPIA Publication 273, December 1975.
 6. T.L. Brosseau and J.R. Ward, "Reduction of Heat Transfer in 105mm Tank Gun by Wear-Reducing Additives", BRL Memorandum Report No. 2698, November 1976. (AD #B015308L)
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 8. R.O. Wolff, "Reduction of Gun Erosion, Part II. Barrel Wear-Reducing Additive", Picatinny Arsenal Technical Report No. 3096, August 1963.
 9. H. Yamazaki, A. Izumi, and T. Hirasawa, "Study on Additives for Reducing Gun Barrel Erosion (8th Report)", JDA Technical Research and Development Institute Report, July 1976.

TABLE II. Composition and Physical Dimensions of Propellants
With Japanese Additives

<u>Composition, Percent by Weight</u>		
	<u>Lot No. AC-E-1A</u>	<u>Lot No. AC-E-1B</u>
Nitrocellulose (12.6%)	28.33	28.92
Nitroglycerin	22.31	21.60
Nitroguanidine	47.59	47.61
Ethylcentralite	1.49	1.59
Cryolite	0.28	0.28
Graphite (added over)	0.08	0.09
DL- α -alanine (added over)	-----	-----
Calcium Pyroglutamate (added over)	5	5
Volatile Matter	0.21	0.40
 <u>Physical Dimensions, mm</u>		
Length	14.87	14.75
Outer Diameter	6.30	6.05
Bore Diameter	0.79	0.84
Web Thickness	1.10	1.03
Shape	7-hole MP	7-hole MP

Cellulose nitrate used in military propellants is a mixture of polymers of varying chain lengths; hence, a correct chemical formula for it contains fractional numbers of atoms. The present calculations circumvent this difficulty by the arithmetical artifice of considering the cellulose nitrate in each propellant as being made of the appropriate mixture of cellulose dinitrate and cellulose trinitrate. No physical reality is thereby implied. The total amount of cellulose nitrate is readily obtained by adding the amounts of dinitrate and trinitrate, although the percentage of nitrogen in the actual cellulose nitrate must be computed.

It is traditional for propellant description sheets to present the composition of the final mix in terms of principal ingredients and "added" materials. The principal ingredients are always specified on a dry basis and in amounts that add to 100 parts (by weight). The "added" materials are specified as parts per hundred added afterward; the figures on the description sheet add to a number greater than 100. Also, the varying amounts of residual solvent and impurities are given as percentages of the final material as determined by analysis after manufacture. Hence, the percentages of each ingredient in the propellant used to compute thermochemical properties are different from the percentages given on the description sheets. These arithmetical adjustments are absolutely necessary; no meaningful comparison between different formulations can be made without them. Table III lists the percentages

of each ingredient used in the thermochemical calculations. The heats of formation of alanine and calcium pyroglutamate were computed earlier by Freedman.¹⁰ These heats of formation are -563.5 kJ/mole and 1572 kJ/mole for alanine and calcium pyroglutamate, respectively.

TABLE III. Percentage by Weight of Constituents in Japanese Propellant Used in BLAKE Computations

Constituent	AC-E-1A	AC-E-1B	AC-E-1A (minus alanine)
Cellulose Dinitrate	13.365	14.041	14.367
Cellulose Trinitrate	13.185	12.556	13.881
Nitroglycerin	21.130	20.620	22.245
Nitroguanidine	45.070	45.452	47.452
Ethyl Centralite	1.410	1.518	1.486
Cryolite	0.265	0.268	0.279
Graphite	.080	.091	.080
Alcohol	.210	.404	.210
Alanine	5.000	-----	-----
Calcium Pyroglutamate	-----	5.051	-----

The results of the BLAKE code computations are listed in Table IV. One computation was done with the alanine removed from the Japanese propellant in order to show how the alanine alters the thermochemistry. Calculations for M30 propellant are also tabulated using values from an earlier computation with the BLAKE code.¹¹ The results show the alanine and calcium pyroglutamate lower the propellant flame temperature and impetus relative to M30 propellant. Hence, one is not surprised that the Japanese propellants reduced the erosion rate of the 90mm tank cannon, since lower flame temperature propellants produce less erosion as exemplified in Figure 1. One would expect the muzzle velocity and chamber pressure to be lower with the Japanese propellants unless the charge mass was increased to compensate for the reduced impetus.

A similar experiment was done in the US which illustrates the problem with putting additives in the propellant.¹² The standard US wear-reducing liner is a blend of titanium dioxide and wax that is coated on a rayon liner which is then cemented to the cartridge case wall. A test was done in the 105mm M68 tank cannon firing M456 HEAT

10. E. Freedman, BRL Memorandum Report in preparation.
11. R. Geene, et al, "Erosivity of a Nitramine Propellant", Proceedings of the 1978 JANNAF Propulsion Meeting, February 1978.
12. P.R. Grepps, "Component Development Tests of Cartridge, 105mm HEAT M456, With Titanium Dioxide Additive to Determine Gun Tube Wear Characteristics", DPS Report 978, June 1963.

TABLE IV. Summary of Propellant Thermochemical Properties*

<u>Propellant</u>	<u>Adiabatic Flame Temperature, K</u>	<u>Impetus, J/g</u>	<u>Ratio of Specific Heats</u>	<u>Molecular Weight of Gases, kg/kg-mole</u>
M30	3040	1088	1.238	23.2
AC-E-1A (Minus Alanine)	3016	1078	1.241	23.3
AC-E-1A	2691	1000	1.252	22.4
AC-E-1B	2632	956	1.252	22.9

* - For a density of loading of 0.2 g/cm^3 .

cartridges to see if TiO_2 blended directly into the propellant would be as effective as the TiO_2 /wax liner. Propellant lots were prepared with two and five percent by weight TiO_2 ; the propellant description is given in Table V. It was learned that only 0.323 kg of extra propellant could be added to the M456 cartridge. The results of the test are listed in Table VI where one sees that the muzzle velocity and chamber pressure are reduced for the five percent TiO_2 . One sees that the erosion rate of the M68 tank cannon is reduced substantially with the five percent TiO_2 .

TABLE V. Propellant Description for TiO_2 -Impregnated M30 in Percent by Weight

Constituent	PE-100-6	PE-100-7
Nitrocellulose	27.92	28.06
Nitroglycerin	22.23	22.76
Nitroguanidine	48.03	47.33
Ethyl Centralite	1.58	1.60
Cryolite	0.24	0.25
Titanium Dioxide (added)	1.95	4.90
Total Volatiles	0.33	0.49
Graphite	0.09	0.09
Ash	2.31	1.90
Dust	0.01	0.02

BLAKE code calculations were made for the TiO_2 -impregnated propellant to see how the flame temperature and impetus change with the addition of TiO_2 . The library for the BLAKE thermochemical program does not contain data for titanium compounds, so ZrO_2 was used in place of TiO_2 in the computations. Since the overall level of the additive is small, and there is no significant chemical interaction for either element, the net effect on the computations is a small one caused by the difference in atomic weights between titanium and zirconium. To make the substitution more realistic, the enthalpy of formation of TiO_2 was used for ZrO_2 .

The TiO_2 -impregnated propellants differ from M30 in one interesting aspect, the surprisingly large amount of ash found in them. Even if one assumes that this ash is absolutely chemically inert, its presence cannot be ignored, because it displaces proportionate amounts of the more energetic ingredients, especially nitroglycerin and nitroguanidine. Its presence in the computations was simulated by using a corresponding amount of alundum, Al_2O_3 , but with a more negative enthalpy of formation than real alundum has in order to make the "ash" less reactive. The results of the BLAKE computations are listed in Table VII where one sees the lower flame temperature and impetus for the TiO_2 (and ash)-impregnated M30 propellant.

TABLE VI. Results of Modifying M30 Propellant by Addition of TiO_2

<u>Propellant</u>	<u>Charge Mass, kg</u>	<u>Chamber Pressure, MPa</u>	<u>Muzzle Velocity, m/s</u>	<u>Wear Rate, mm/round</u>	<u>Rounds Considered</u>
M30 (RAD-64550)	5.205	421	1185	1.4 ^a 0.18 ^c	1000 ^b 507
2% TiO_2 (PE-100-6)	5.528	415	1183	1.0	99
5% TiO_2 (PE-100-7)	5.528	351	1110 ^d	0.10	100

a - No additive.

b - Total rounds fired in six tubes.

c - TiO_2 -wax additive.

d - About one-half the muzzle velocity drop could be recovered by tailoring the web to produce same peak chamber pressure as M30. The erosion rate will be slightly higher as well.

TABLE VII. Thermochemical Computations for TiO_2 -Impregnated M30*

<u>Propellant</u>	<u>Adiabatic Flame Temperature, K</u>	<u>Impetus, J/g</u>	<u>Ratio of Specific Heats</u>	<u>Molecular Weight of Gases, kg/kg-mole</u>
M30	3040	1088	1.238	23.2
2% TiO_2 (PE-100-6)	2749	944.6	1.246	23.2
5% TiO_2 (PE-100-7)	2608	875.1	1.249	23.2

* - For a loading density of 0.2 g/cm^3 .

IV. DISCUSSION

The net effect of decreasing the amounts of energetic ingredients and adding filler, either alanine, calcium pyroglutamate, or TiO_2 , with large negative enthalpies of formation is that the final composition has a computed enthalpy of formation of approximately -2.1 kJ/g compared to only -1.2 kJ/g for the M30 formulations usually studied. As seen from the computations in Tables IV and VII, this results in a flame temperature some 300K to 400K lower for the present formulations. All of the computations can be easily summarized. When an inert ingredient is added homogeneously to a propellant, the flame temperature of that propellant is lowered. There is no exception to this conclusion. The word homogeneously is emphasized because that is the only case considered by the code. If the additive were wrapped around the outside of the propellant, the resulting temperature would be quite different near the tube wall, and beyond the computational resources of the code used here. But such was not the case in any of the experiments that inspired this set of computations.

The chemistry of the organic additives is completely described by the code, and needs no further discussion. If either alanine or calcium pyroglutamate have any effectiveness as erosion reducers, it is because their presence in a propellant lowers the flame temperature. This same result can be achieved more readily by decreasing the amount of nitroglycerin in the propellant formulation.

V. CONCLUSIONS

1. Addition of alanine or calcium pyroglutamate to M30 propellant reduces the adiabatic flame temperature and impetus. The reduction in erosion seen in the 90mm M41 tank gun could be achieved more easily by reducing the nitroglycerin or nitroguanidine content of M30 propellant.

2. The Japanese inventors overestimated the improvement in tube life for the 90mm tank cannon using M30 propellant with either alanine or calcium pyroglutamate. By separating the number of rounds needed to remove chromium plating from the erosion of the underlying steel, one finds the tube life was increased from 250 to 400 rounds, not the threefold increase to which the Japanese extrapolated by dividing wear by total rounds fired.

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